

Investigation on syngas production via biomass conversion through the integration of pyrolysis and air–steam gasification processes



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ARTICLE INFO

Article history:

Received 18 February 2014

Accepted 23 July 2014

Keywords:

Renewable energy

Biomass

Pyrolysis

Gasification

Syngas

Hydrogen

ABSTRACT

Fuel production from agro-waste has become an interesting alternative for energy generation due to energy policies and greater understanding of the importance of green energy. This research was carried out in a lab-scale gasifier and coconut shell was used as feedstock in the integrated process. In order to acquire the optimum condition of syngas production, the effect of the reaction temperature, equivalence ratio (ER) and steam/biomass (S/B) ratio was investigated. Under the optimized condition, H₂ and syngas yield achieved to 83.3 g/kg feedstock and 485.9 g/kg feedstock respectively, while LHV of produced gases achieved to 12.54 MJ/N m³.

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1. Introduction

The rapid population growth, economic crisis, world energy consumption and impending depletion of fossil fuels make findings related to alternative and renewable fuel resources essential. Energy resources such as coal, oil and natural gas are being consumed at an accelerated rate and their depletion in the next few decades is one of global concern. Reports estimated that some of the oil rich countries would fail to meet the world energy demand in the next few decades [1]. Meanwhile the world CO₂ emission which is the main cause of global warming has increase by 3% and reached an all-time high of 34 billion tons in 2011 [2]. These figures have led to the Doha amendment to the Kyoto Protocol that states industrialized countries must reduce their greenhouse gas emissions to 30% by 2020 [3]. Accordingly, biomass as an abundant feedstock resource of renewable energy has significant economic and environmental potential to produce syngas value added fuel by a conversion process, while at the same time addressing emerging issues concerning global energy needs and carbon emission.

Syngas is one of the gasification products and emerging alternative fuels that could derive from biomass. The energy density of syngas is about 50% of natural gas and is mostly suited for transportation fuel application and chemical production. The syngas

produced can be directly fed to SOFC units, or it can be further transformed into hydrogen through specific CO clean-up processes for feeding PEMFC units for the production of clean energy.

Thermochemical conversion technologies are promising options for the utilization of biomass as a source of energy. The pyrolysis process has proven to be an appropriate technology for biomass conversion to value added products and has attracted the consideration of many researchers; especially flash pyrolysis which is a well known process for converting biomass to light and heavy hydrocarbons is particularly interesting. Many pyrolysis reactors and processes have been developed in the last decade to derive gas, char and bio-oil from biomass [4–9]. Besides the pyrolysis process, gasification presents an attractive alternative for syngas production. Several studies have been performed to investigate the effects of various parameters on gasification and utilization of biomass as a source of renewable energy [10–17].

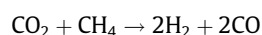
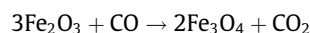
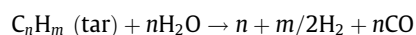
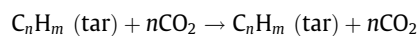
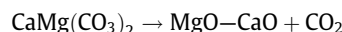
Since the tar removal and conversion of hydrocarbons to syngas increases the overall efficiency and economic viability of biomass gasification, catalysis has become an important parameter in the gasification process and is able to elevate the syngas yield. There are several types of catalysts; the alkali metal catalyst that is considered to be a tar eliminator and catalyst for upgrading the product gas [18–22]. The nickel catalyst has the highest attention of researchers and is widely used in industrial processes in order to hot gas cleaning in biomass gasification [23–27]. The dolomite with general formula of MgCO₃·CaCO₃ is a capable catalyst for

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upgrading the syngas quality for hot gas cleaning and it may be used as a primary, dry-mix or either downstream catalyst in the gasifier. As it is an inexpensive disposable catalyst that can significantly convert and reduce the tar content of produced gas, it has attracted the attention of many researchers [28–34].

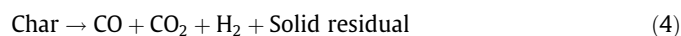
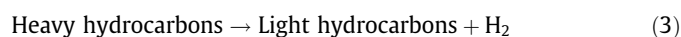
The reactions that are involved in tar conversion through the dolomite catalyst are considered below;



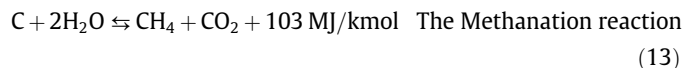
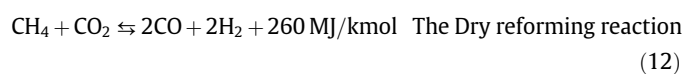
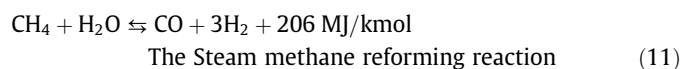
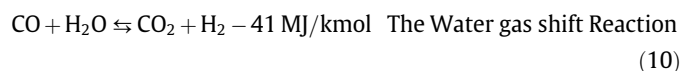
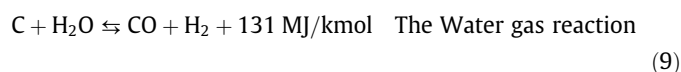
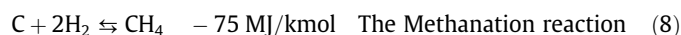
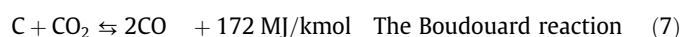
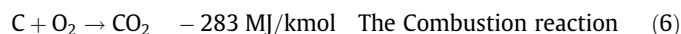
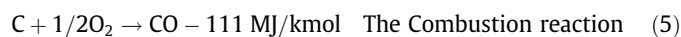
The gasifying agent has a significant impact on the gasification reactions. Air, as a common gasifying agent which contains a large quantity of nitrogen, causes the heating value of the syngas produced to decrease. Pure oxygen and steam are valid alternatives as they increase the heating value; however these are not economical as high operating costs are involved in purifying oxygen and super heating steam [11,12]. By using a mixture of air and steam, the quality of the syngas and overall syngas yield will be enhanced without the high operating costs.

The concept of combination and integration of the pyrolysis and gasification process has the purpose of producing a high quality gas, in terms of low tar content, and also shift the reactions from exothermic to endothermic, which result in an enhancement of the syngas yield and also the heating value of the gases produced [35–39].

Biomass conversion to syngas in pyrolysis and gasification processes occurs under the main steps that are indicated by the reactions below:



The formation and consumption of CO, CO₂, CH₄, H₂ and steam through the pyrolysis and gasification processes can be studied via the given reactions in (Eqs. (5)–(13)) that occur at various degrees.



Moreover, Warnecke [40] claimed that the design of the gasifier has a major influence on product gas composition. Additionally, the design of a gasifier can cause the same feedstock to give different calorific values of a product. Among all designs, depending on the size and density of the fuel, the fluidized bed gasifier has been shown to be a versatile technology that makes it capable for biomass conversion with higher efficiency in terms of heat and mass transfer and also high reaction rates due to intensive mixing in the bed. In this study more emphasis has been given to the optimization condition of integrated pyrolysis and gasification process in fluidized bed gasifier in order to biomass conversion into value-added syngas fuel.

2. Experimental section

2.1. Feedstock and experimental setup

Coconut shell is a well known biomass waste due to its high calorific value and fixed carbon. Coconut is the fourth important industrial crop in Malaysia in terms of total planted area recorded in 2012 and its production was about 74×10^6 nuts through the country [41]. The samples were pulverized into powder and sieved into a specific particle size in range of 100–300 μm . The proximate and ultimate analyses of feed stock are reported in Table 1.

The schematic diagram of the experimental facility used in this study is shown in Fig. 1. The setup consists of a cylindrical reactor made of (Inconel 625) with the following design; pyrolysis zone: i.d. of 25 mm and height of 250 mm, gasification zone: i.d. of 55 mm and height of 450 mm and catalytic bed zone: i.d. of 55 mm and height of 300 mm. Two individual electrical heaters equipped with a temperature indicator controller (TIC) were installed on the pyrolysis and gasification zones of the gasifier. Three thermocouples were set across the gasifier, dense bed, gasification zone and catalytic bed. The flexible design of the gasifier leads to saving energy and enhancing the syngas production.

2.2. Experimental procedures

An experimental schedule was performed to analyze the individual effect of the main parameters governing the produced syngas quality and process efficiency. At the startup of the experiment, the gasifier was charged with 100 g silica sand with U_{mf} of 0.18 m/s as bed material, which enabled to stable fluidization and better heat transfer. The feedstock with feeding rate of 0.78 kg/h were introduced to the pyrolysis zone and fluidized by 10 L/min of nitrogen injection. Super heated steam at 270 °C and pre-heated air with a flow rate of 1.1 N m³/h were applied as gasifying agent in the gasification zone. The gas produced from char gasification was introduced into the catalytic bed while there was a secondary injection of 0.5 kg/h of super heated steam that enhanced the hydro-cracking process. The gas stream was passed through the water-cooled heat exchanger, followed by a condenser trap and a cyclone. Online Gas Chromatography (GC) (Agilent 7890A) equipped with a Thermal Conductivity Detector (TCD) was employed to analyze the permanent gases H₂, CO, CH₄ and CO₂ produced.

3. Results and discussion

3.1. Effects of temperature

3.1.1. Pyrolysis zone

Temperature profile is one of the crucial parameters in overall biomass conversion and has a major influence on the final product

Table 1
Proximate and ultimate analysis of coconut shell.

Proximate analysis (wt% wet basis)		Ultimate analysis (wt% dry basis)				
Moisture content	8.55	C	H	O	N	S
Volatile matter	52.56	50.2	5.40	43.4	0.94	0.06
Fixed carbon	26.45	Density (kg/m ³)				661
Ash	12.44	HHV (MJ/kg)				21.50

composition. To verify the importance of the pyrolysis zone temperature on gas composition and related LHV, a series of experiments in the range of 400–650 °C were performed and obtained the results presented in Table 2.

$$\text{LHV (MJ/N m}^3\text{)} = ([\text{H}_2] * 107.98 + [\text{CO}] * 126.36 + [\text{CH}_4] * 358.18 + [\text{C}_2\text{H}_2] * 56) / 1000$$

The results obtained confirmed that the increasing temperature in the pyrolysis zone of the gasifier causes the syngas production to be enhanced up to a maximum fraction of 33.12 vol% and 16.57 vol% for H₂ and CO respectively. This is due to enhance the hydro-cracking of heavy hydrocarbons and further conversion through reforming reactions that subsequently contribute to the syngas production with a maximum syngas yield of 382.4 g/kg feedstock [37,38,42].

3.1.2. Gasification zone

Figs. 2 and 3 represent the gas composition and product yield achieved by varying the gasification zone temperature in the range of 750–1100 °C while ER, S/B ratio and pyrolysis zone temperature were kept constant at 0.24, 0.64 and 500 °C respectively. In this study the gasification efficiency was reflected by the carbon conversion which achieved up to a maximum of 84.1% and was calculated by,

$$X_C (\%) = (12Y(\text{CO}\% + \text{CO}_2\% + \text{CH}_4\% + 2 * \text{C}_2\text{H}_4\%)) / (22.4 * \text{C}\%) * 100$$

$$\text{ER} = \left(\frac{\text{the actual oxygen fuel ratio}}{\text{the oxygen fuel ratio for complete combustion}} \right)$$

$$\text{EY}_{\text{syngas}} = \left(\frac{\left(\frac{m_{\text{H}_2} + m_{\text{CO}}}{m_{\text{total gas}}} \right) * \text{DGF}}{m_{\text{total biomass}} + m_{\text{st. steam}}} \right)$$

DGF: dry gas flow rate

The tendency of high temperature favored syngas production has been widely reported [43–46]. This phenomenon could be due to the contribution of the water–gas reaction, the steam methane reforming and the dry reforming reactions which become dominant during the gasification process at high temperatures and consequently cause an increase in the amount of total syngas in accordance with the endothermic char gasification [46]. As observed, gasification reactions favored high temperature and the process was influenced by endothermic reactions. Hence, increasing the gasifier temperature up to 1100 °C enhances the H₂ and CO production to a maximum value of 43.8 vol% and 36.8 vol% respectively. Furthermore, the syngas yield increased from 177.17 to 409.9 g/kg feedstock in the range of the applied temperature. On the other hand, CH₄ and CO₂ consumption progressed and these fractions decreased to 9.4 vol% and 10.1 vol% respectively due to reforming and boudouard reactions. The H₂/CO ratio of 1.19 and CO/CO₂ of 3.63 achieved at 1100 °C indicate the maximum of these ratios. These observations are in accordance with the results obtained by previous studies [47–50].

3.2. Effect of catalyst

Table 3 presents the results of dolomite application in the integrated pyrolysis and gasification process. For the tests performed

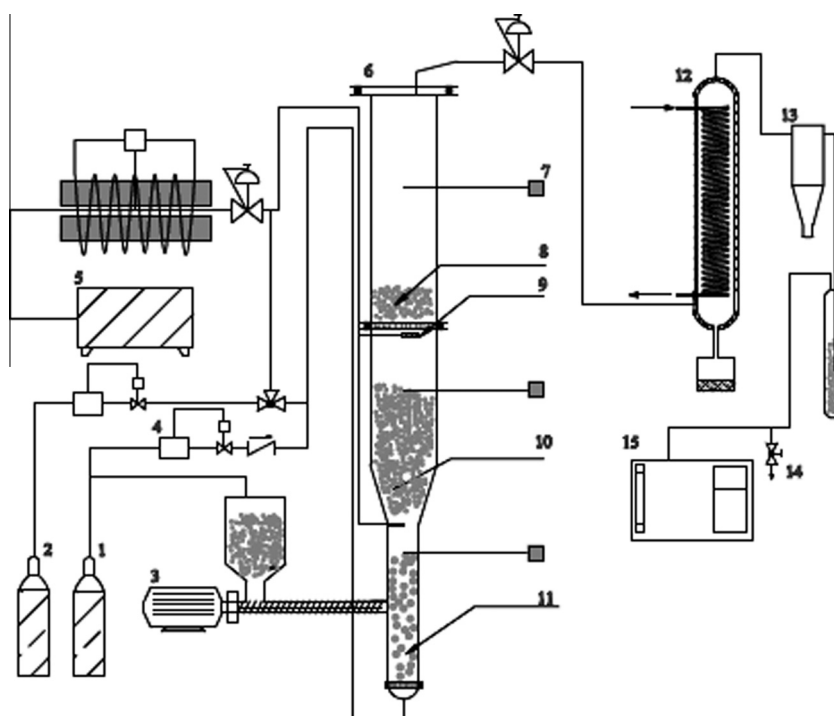
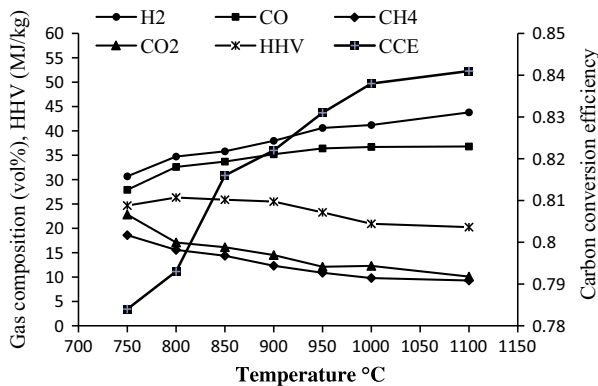
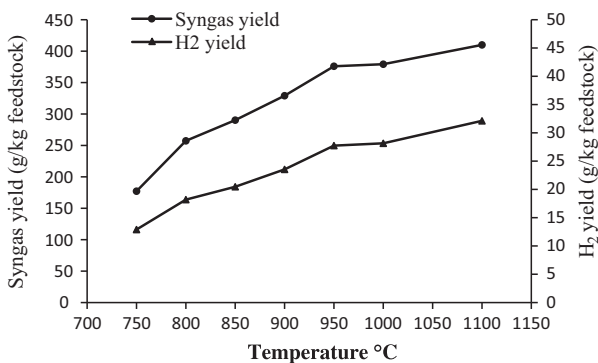


Fig. 1. Process flow diagram of bench scale integrated pyrolysis and air–steam catalytic gasification setup. 1. Nitrogen gas, 2. Air, 3. Feeder, 4. Mass flow controller (MFC), 5. Boiler, 6. Gasifier, 7. Thermocouple, 8. Catalytic bed zone, 9. Steam injector, 10. Gasification zone, 11. Pyrolysis zone, 12. Condenser, 13. Cyclone, 14. Off line gas sampling, 15. Gas chromatograph.

Table 2

Effect of pyrolysis zone temperature on product gas and syngas yield.

Parameters	R1	R2	R3	R4	R5	R6	R7
Feed rate (kg/h)	0.78	0.78	0.78	0.78	0.78	0.78	0.78
Gasification reaction temperature (°C)	950	950	950	950	950	950	950
Pyrolysis reaction temperature (°C)	400	420	450	500	550	600	650
S/B (kg/kg)	–	–	–	–	–	–	–
H ₂ (vol%)	23.66	24.85	26.68	28.67	29.86	31.13	33.12
CO (vol%)	12.21	12.34	14.27	15.42	15.76	16.12	16.57
CH ₄ (vol%)	12.24	11.27	10.58	10.39	9.57	9.42	8.11
CO ₂ (vol%)	8.61	8.32	7.67	7.35	7.12	6.9	6.45
LHV (MJ/N m ³)	8.48	8.28	8.47	8.46	8.64	8.77	8.57
Dry gas flow rate (kg/h)	0.45	0.48	0.52	0.57	0.6	0.61	0.61
Syngas yield (g/kg feedstock)	206.4	228.6	277.9	320.7	349.5	362.4	382.4
H ₂ yield (g/kg feedstock)	25.1	28.76	32.74	37.61	41.66	43.94	47.78
H ₂ /CO	1.88	2.01	1.87	1.86	1.89	1.93	1.99

**Fig. 2.** Effect of temperature on gas composition.**Fig. 3.** Effect of temperature on syngas and H₂ yield.

through the application of dolomite, H₂ and CO content increased to 51.48 vol% and 26.01 vol% respectively, while the CO₂, CH₄ and C_nH_m production was reduced. Furthermore, the use of steam in the presence of dolomite enhanced the H₂ and syngas yield to a maximum value of 30.86 g/kg feedstock and 352.25 g/kg feedstock respectively. In addition, the H₂/CO ratio increased from 1.64 to 2.1. Through the application of dolomite, the tar conversion reached a maximum efficiency of 85.06% that this phenomenon may explain due to the existence of iron component (0.7 wt% Fe₂O₃) in structure of dolomite as it is a favorable metal for the steam reforming and water gas shift reactions [24,29]. According to Rownaghi and Huhnke [51], the CaO and MgO has the ability to convert the tar up to 48% and 42% respectively. Additionally, they reported that, by adding the NiO to raw MgO plus CaO (CaO/MgO/NiO), the activity of tar conversion of these catalysts could rise from below 50% up to 100% at 700 °C.

3.3. Effect of equivalence ratio (ER)

In this research the equivalence ratio varied from 0.14 to 0.27 through changes to the air flow rate under the three different temperatures of 900 °C, 950 °C and 1000 °C, while other conditions were held constant. The test results of varying ER were reported in Fig. 4a–c.

By increasing ER from 0.14 to 0.24 the total syngas concentration was enhanced and reached up to maximum values of 42.02 vol%; 44.62 vol% and 45.18 vol% for temperatures of 900, 950 and 1000 °C respectively. At ER: 0.24 the char gasification reached maximum efficiency and caused H₂ and CO to achieve their maximum value in the range of the applied temperature. On the other hand, CO₂ and CH₄ content reduced through boudouard and reforming reaction. Further increasing the ER could lead to the reduction of total syngas concentration due to excess air and would also cause a decline of temperature in the oxidation zone. Furthermore, Lv et al. [52] stated that, the ER not only represents the quantity of oxygen introduced into the gasifier, but it also indicates the boundary of gasification and combustion inside the gasifier.

3.4. Effect of steam/biomass ratio (S/B)

The S/B ratio is an influential parameter on the gasification process and is important to find the existing optimum point in the conversion process since too large an S/B ratio does not always favor the syngas production and is not cost effective. Figs. 5 and 6 represent the effect of the S/B ratio over the range of 0.16–3.1 on syngas production and gasification efficiency.

Through enhancing the conversion reactions by a secondary injection of steam in the catalytic bed, Reforming reactions, water gas shift and char gasification became governed reactions in the overall biomass conversion process and caused an increase in the syngas production. Over the range of the applied S/B ratio, the syngas yield achieved up to a maximum value of 490.74 g/kg-feedstock with composition of 64.62 vol% H₂ and 22.28 vol% CO. On the other hand, CH₄ and CO₂ content had a decreasing trend due to reactions (7), (11) and (12). The H₂/CO ratio increased to 2.9 and also CO/CO₂ ratio showed an increasing trend from 1.51 to 4.14. The LHV decreased in the range of S/B ratios to 12.56 MJ/N m³ which indicated a dominance of steam methane reforming while the carbon conversion efficiency (CCE) increased to 89.2% due to the enhancement of the char gasification.

3.5. Optimization condition

The process optimization was based on the syngas and hydrogen yield. Referring to obtained results the temperature parameter

Table 3

Effect of dolomite catalyst on syngas quality.

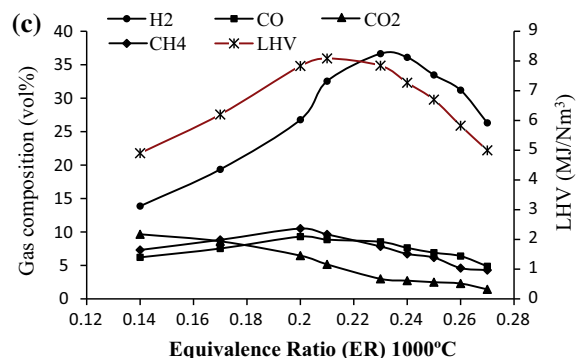
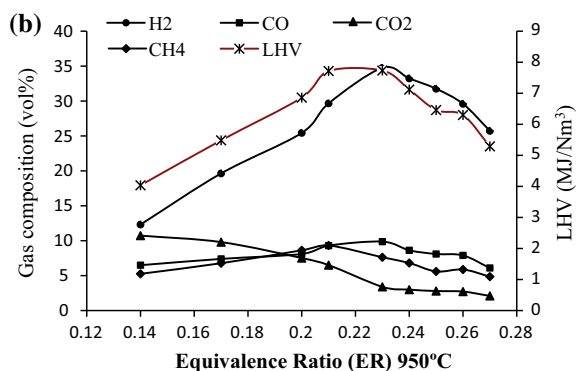
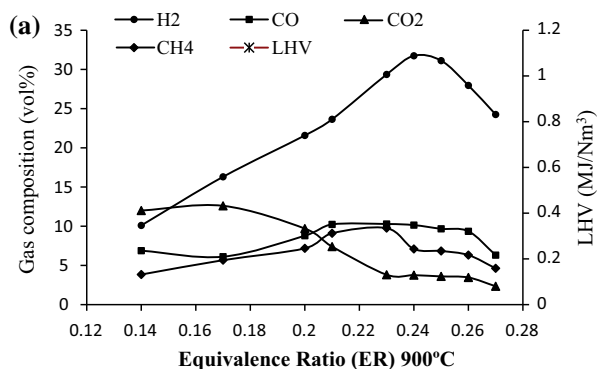
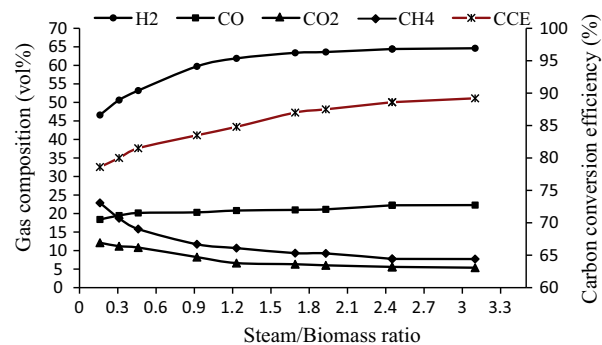
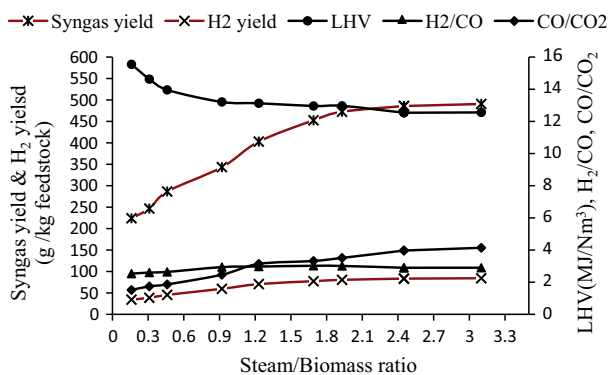
P. (°C)	G. (°C)	C. (°C)	S/B ratio	C/B ratio	H ₂ (vol%)	CO (vol%)	CO ₂ (vol%)	CH ₄ (vol%)	Tar (g/N m ³)	Tar con. (%)	H ₂ yield	Syngas Yield	H ₂ /CO
500	850	730	2	–	38.16	23.18	17.24	21.42	7.63	–	22.52	214.12	1.64
500	850	730	2	1	46.57	24.58	12.64	16.21	2.36	69.1	26.19	297.42	1.89
500	850	730	2	1.5	50.62	24.15	10.56	14.67	1.37	82.04	28.73	330.31	2.1
500	850	730	2	2	51.48	26.01	10.33	12.18	1.14	85.06	30.86	352.25	1.98

P: pyrolysis zone temperature, G: gasification zone temperature, C: catalytic bed.

Table 4Syngas and H₂ yield at optimum conditions.

P. (°C)	G. (°C)	S/B	ER	C/B (kg/kg)	Gas flow rate (kg/h)	CCE (%)	Total syngas (vol%)	H ₂ yield	Syngas yield	H ₂ /CO	LHV (MJ/N m ³)
500	950	1.69	0.24	1.5	0.7	93.6	85.37	77.3	452.6	3.31	12.95
500	950	1.93	0.24	1.5	0.72	93.9	85.74	80.4	472.5	3.01	12.97
500	950	2.45	0.23	1.5	0.74	94.1	86.65	83.3	485.9	2.89	12.54
500	950	3.1	0.23	1.5	0.74	95.8	86.9	84.2	490.7	2.9	12.55

P: pyrolysis zone temperature, G: gasification zone temperature, C: catalyst.

**Fig. 4.** (a–c) Effect of equivalence ratio (ER) on gas composition.**Fig. 5.** Effect of S/B ratio on gas composition. ER: 0.24 gasification zone: 950 °C pyrolysis zone: 500 °C.**Fig. 6.** Effect of S/B ratio on syngas yield, H₂ yield, H₂/CO, CO/CO₂ and LHV. ER: 0.24 gasification zone: 950 °C pyrolysis zone: 500 °C.

optimized at 500 and 950 °C for pyrolysis and gasification zone respectively, equivalency ratio (ER) at 0.23–0.24 and the optimum S/B ratio found in range of 1.9–2.5 (Table 4).

4. Conclusion

In the present study, under the optimized condition the syngas yield achieved a value of 485.9 g/kg feedstock with a fraction of

64.42 vol% of H_2 and 22.23 vol% of CO and H_2 yield achieved to 83.3 g/kg feedstock. Among the variable parameters, the temperature appeared to have the most pronounced effect on syngas production. The ER had complex effects and obtained results confirmed an optimal value existed for this factor, which was different according to different operating parameters. The S/B ratio revealed that there is an optimum value of this parameter according to the composition of feedstock. The injection of steam to the catalytic bed caused a significant reduction of tar and increases the efficiency of reforming reactions.

Future work can extend the study by conducting research on the application of different types of premixed catalyst with biomass and testing within the existing integrated process for a study on the further enhancement of the syngas yield.

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